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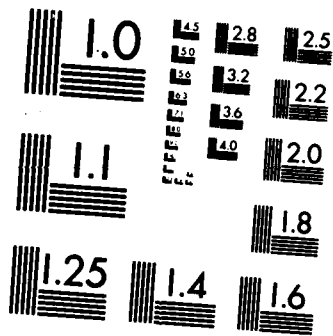
TRANSITION DIPOLE-SOLVENT INTERACTION IN  
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TRANSITION DIPOLE-SOLVENT INTERACTION IN PHOTOIONIZATION

by

Paul Delahay and Andrew Dziedzic

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# TRANSITION DIPOLE-SOLVENT INTERACTION IN PHOTOIONIZATION

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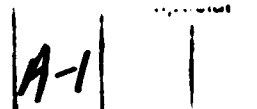
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The interaction between solvent molecules and transition dipoles results in a measurable nonequilibrium electronic contribution to the energetics of photoionization in the case of significant dielectric dispersion of the solvent at the prevailing photon energy. Earlier theoretical work on the resulting dispersion shift is justified on the basis of transition dipole-solvent interaction. Additional experimental evidence is presented.

## 1. Introduction

Spectroscopic transitions from one bound state to another bound state involve a transition dipole which oscillates at or very near the frequency of the incident radiation [1]. Such an oscillating transition dipole is also involved in transitions to the continuum [2], but there is, of course, in that case ultimate separation of the emitted electron from its parent atomic or molecular system. In solutions, the transition dipole interacts with surrounding solvent molecules in a process which is similar to solvation of a dipole or, in the limit of complete charge separation, to ionic solvation except that nuclear motion is not involved. The response of the solvent is determined by its dielectric properties at the frequency at which the transition dipole oscillates, that is, by the real and imaginary optical dielectric constant of the solvent at or near the frequency of radiation. The complex dielectric constant is a function of photon energy because of dispersion and consequently the energy for the transition dipole-solvent interaction varies with the energy of the incident radiation.



In the absence of dispersion, the free energy for transition dipole-solvent interaction depends on the limiting value  $\epsilon_{op}^0$  of the optical dielectric constant (in the near infrared range). This free energy of interaction for the dielectric constant  $\epsilon_{op}^0$  is part of the difference in free energy between the initial and final states of the photoionization process. The difference between the free energies of transition dipole-solvent interaction for the dielectric constant at the prevailing photon energy and  $\epsilon_{op}^0$  therefore represents a nonequilibrium electronic contribution to the free energy of photoionization. The expression dispersion shift is suggested for this nonequilibrium electronic contribution.

Two main questions arise about the dispersion shift: (i) What experimental evidence is there which supports the introduction of a dispersion shift depending on photon energy as part of the photoionization energy in liquids and solutions? (ii) How can the dispersion shift be calculated? Experimental evidence was presented in earlier paper [3] and theory was developed in [4]. Additional evidence is reported in the present Letter and the previous theory is given further conceptual support.

## 2. Experimental evidence for the dispersion shift

Strong support for the occurrence of transition dipole-solvent interaction is available from experiments on photoelectron emission by liquids and solutions [5]. In these experiments, normally incident VUV radiation strikes the liquid surface and emitted photoelectrons are collected on a planar grid electrode parallel to the liquid surface. An emission yield,  $Y$ , is calculated as the number of collected electrons per incident photon. In the absence of a dispersion effect,  $Y$  is proportional to  $(E - E_t)^2$ , where  $E$  is the photon energy and  $E_t$  the threshold energy for the species being photoionized. This quadratic emission law holds at photon energies at least a few tenths of an

electronvolt higher than  $E_t$ . Therefore, a plot of  $Y^{1/2}$  against  $E$  should be linear, and the derivative  $dY^{1/2}/dE$  should be independent of photon energy in the range of validity of the quadratic law. These conclusions hold if dispersion of the solvent does not affect the variations of  $Y$  with  $E$ . This is the case, for instance, for photoelectron emission by liquid water ( $E_t = 10.06$  eV) because the effect of dispersion on emission for this substance is nearly independent of photon energy in the range of quadratic extrapolation from 10 to 11 eV.

Emission spectra ( $Y$  against  $E$ ) and plots of  $Y^{1/2}$  against  $E$  generally exhibit a fine structure, and the derivative  $dY^{1/2}/dE$  is not independent of  $E$  (fig. 1) above the threshold energy. This effect was first reported for various inorganic anions in aqueous solution [3] but it is of general nature. The functional dependence of  $dY^{1/2}/dE$  on  $E$  (dispersion spectrum) is essentially determined by the solvent and in a minor way by the nature of the substance being photoionized. This is conclusively shown by a series of experiments.

Firstly, dispersion spectra are not the result of an experimental artefact having its origin in the vacuum ultraviolet radiation source since practically the same dispersion spectrum is obtained from 7 to 9 eV (fig. 1) with two sources having significantly different spectral outputs (fig. 2). The difference between the spectra in fig. 1 above 9 eV is caused by the very low output of the argon source in that range and the large error in the normalization of the photon flux.

Secondly, dispersion spectra do not result from an experimental artefact having its origin in the monochromator and optical system since very different spectra are obtained for iodide solutions in water and glycerol (fig. 3). The dispersion spectrum of the iodide solution in glycerol above 9.0 eV is quite

similar to the spectrum for pure glycerol because of dominant emission by the solvent. In a separate experiment, water vapor was allowed to be absorbed by the iodide solution in glycerol in the emission chamber. The dispersion spectrum evolved toward the spectrum obtained with aqueous solutions of iodide, as the water was increasingly absorbed by the glycerol.

The dispersion spectrum for a given solvent is essentially independent of kinetic energy of quasifree electrons produced by photoionization. This was shown in ref. [3] for aqueous solutions of 17 inorganic anions with threshold energies ranging from 7.2 to 8.9 eV.

### 3. Theoretical considerations on the dispersion shift

Two limiting cases may be considered in the calculation of the dispersion shift:

(i) One assumes complete separation of the emitted electron from its parent ion or molecule in solution. This approach was developed recently [4], and a detailed calculation was performed of the dispersion shift. A discrete model of solvent molecules was used for the inner-sphere region around the photoionized species, whereas the outer-sphere region was treated in terms of a continuous medium. Both transparent and absorbing solvents were considered.

The dispersion shift was calculated on the ad hoc assumption that the ionic field changes on a time scale corresponding to the radiation frequency. The central idea of the present paper, namely that of transition dipole-solvent interaction, provides the conceptual justification for the calculation in [4]. Only the change in the ionic field from the initial to the final state of photoionization was considered in [4], and no attempt was made to follow the time-evolution of the transition dipole-solvent interaction. Thus, the matrix element for absorption refers only to the initial and final states, and the absorption of a photon is accounted for by the annihilation operator acting in occupation space [6].



The theory of [4] accounts very well for the functional dependence on photon energy exhibited by experimental dispersion spectra. In particular, excellent agreement was obtained between theory and experiment for the photon energies at the 12 extrema exhibited by dispersion spectra of aqueous solutions between 7.2 and 10.4 eV. The effect of a varying screening resulting from a change of electrolyte concentration was also accounted for.

(ii) In the second limiting case, one assumes that the transition dipole is represented by a point dipole. Detailed calculations of the dispersion shift have been performed, and they yield essentially the same functional dependence on photon energy for the dispersion spectrum as the model of complete charge separation. This is easily seen by comparing the expressions for the change of free energy resulting from dispersion for electronic polarization of the outer-sphere region according to the two models. The solvent is taken to be transparent to simplify matters. This change of free energy is

$$\Delta P_{\text{CCS}}^{\text{out}} = Ae^2/2a \quad (1)$$

according to the complete charge separation model, where  $e$  is the electronic charge;  $a = r_c + 2r_w$ , the terms  $r_c$  and  $r_w$  being the crystallographic radii of the ion being photoionized and water, respectively. One has

$$\begin{aligned} A &= [1 - \epsilon_{\text{op}}^{-1}] - [1 - (\epsilon_{\text{op}}^0)^{-1}] \\ &= (\epsilon_{\text{op}}^0)^{-1} - \epsilon_{\text{op}}^{-1} \end{aligned} \quad (2)$$

where  $\epsilon_{\text{op}}^0$  is the limiting value of the optical dielectric constant of the solvent in the near infrared, and  $\epsilon_{\text{op}}$  the optical dielectric constant at the prevailing photon energy.

Conversely,

$$\Delta P_{\text{pd}}^{\text{out}} = B\mu^2/a^3 \quad (3)$$

according to the point dipole model, where  $\mu$  is the transition dipole and

$$B = (\epsilon_{op} - 1)/(2\epsilon_{op} + 1) - (\epsilon_{op}^0 - 1)/(2\epsilon_{op}^0 + 1). \quad (4)$$

Equations (3) and (4) follow directly from the Kirkwood treatment [7] of the solvation free energy of a dipole in a cavity of radius  $a$  surrounded by a continuous medium (cf. eq. (3) in [8]).

One deduces from (1) to (4)

$$\Delta P_{ccs}^{out}/\Delta P_{pd}^{out} = (K/2)(ea/\mu)^2 \quad (5)$$

with

$$K = A/B \\ = [2 + (\epsilon_{op}^0)^{-1}][2 + (\epsilon_{op})^{-1}]/3 \quad (6)$$

The variation of  $K$  with  $\epsilon_{op}$  shows how the two models differ in accounting for the effect of dispersion in the outer-sphere region for a transparent solvent. The range of interest in the present case is  $\epsilon_{op}^0 \leq \epsilon_{op} \leq 2.5$ , and one has  $K = 2.189$  and  $2.050$  for  $\epsilon_{op} = \epsilon_{op}^0 = 1.777$  and  $\epsilon_{op} = 2.5$ , respectively. Thus,  $K$  decreases only by 6.3 percent in a monotonic fashion as  $\epsilon_{op}$  increases from 1.777 to 2.5 whereas  $A$  increases from 0 to 0.163. The two models therefore have essentially the same dependence on dielectric constant although the free energies of polarization have different analytical expressions.

The foregoing conclusion was reached for the outer-sphere region, but it should also be valid for the inner-sphere shell. The treatment of this region involves the polarizability of the solvent in both models, and dispersion is accounted for by the variation of this quantity with photon energy. Only the method of calculating the field acting on the solvent multipoles in the inner-sphere shell varies with the model, and the effect of dispersion is solely contained in the solvent polarizability.

Although the two models predict nearly the same functional dependence of dispersion spectra on photon energies, they yield very different values of the

dispersion shift. Thus, the ratio of (5) is ca.  $540/\mu^2$ , where  $\mu$  is in debyes, and the following data were selected:  $K = 2.05$  ( $\epsilon_{op} = 2.5$ ) and  $a = 4.78 \text{ \AA}$  ( $r_c = 2 \text{ \AA}$ ). Comparison of the theoretical total interaction energy calculated in [4] with experiment in the 7 to 10 eV range shows that the model of complete charge separation yields dispersion shifts which are too high by a factor of ca. three in the absence of ionic screening. Conversely, the point dipole model, in view of the above treatment, should yield dispersion shifts which are too low by ca. one order of magnitude. This is not surprising because the model of a point dipole grossly underestimates the change of electric field around the species being photoionized. Conversely, this change of field is overestimated in the model of complete charge separation because of the finite distance between the charges thus involved. The model of complete charge separation seems preferable because it involves less cumbersome algebra than the treatment of the dipole model would entail without the simplifying assumption of a point dipole.

#### 4. Conclusion

The interaction between solvent molecules and transition dipoles results in a measurable nonequilibrium contribution (dispersion shift) to the energetics of photoionization in the case of significant dielectric dispersion of the solvent at the prevailing photon energy. Two nonequilibrium contributions to the photoionization free energy of liquids and solutions therefore must be considered: (i) the electronic contribution discussed here (dispersion shift) and (ii) the free energy of nuclear reorganization [5]. The functional dependence of the dispersion shift on photon energy is accounted for by theory in agreement with experiment.

#### Acknowledgement

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## Captions to Figures

Fig. 1. Dispersion spectra of 1 M aqueous solution of sodium azide with hydrogen (A) and argon (B) sources having the spectral outputs of fig. 2. Threshold energy of  $N_3^-$ ,  $E_t = 7.4$  eV. Very low output of the argon source above 9.0 eV (dashed portion of B).

Fig. 2. Spectral outputs of hydrogen (A) and argon (B) sources in the experiments of fig. 1. Relative intensities of ca. 0.4 at the normalized highest peaks, the hydrogen source being the stronger.

Fig. 3. Dispersion spectra of 1 M sodium iodide solutions in water (A) and glycerol (B). Dispersion spectrum of pure glycerol (C). Threshold energies,  $E_t \approx 7.4$  eV for  $I^-$  in water and glycerol;  $E_t \approx 8.7$  eV for pure glycerol;  $E_t = 10.06$  eV for pure water. All spectra obtained with hydrogen lamp.

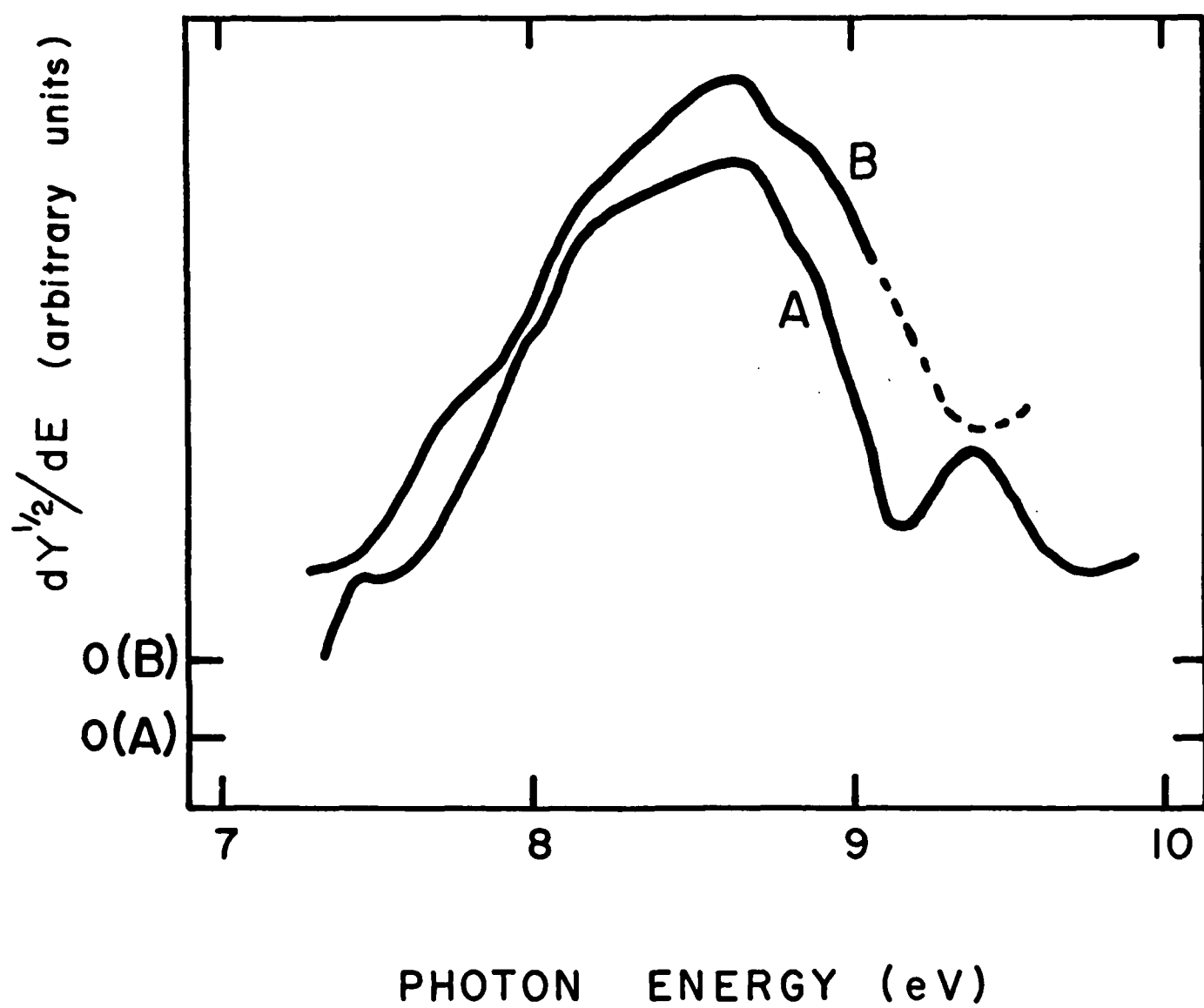


FIG. 1

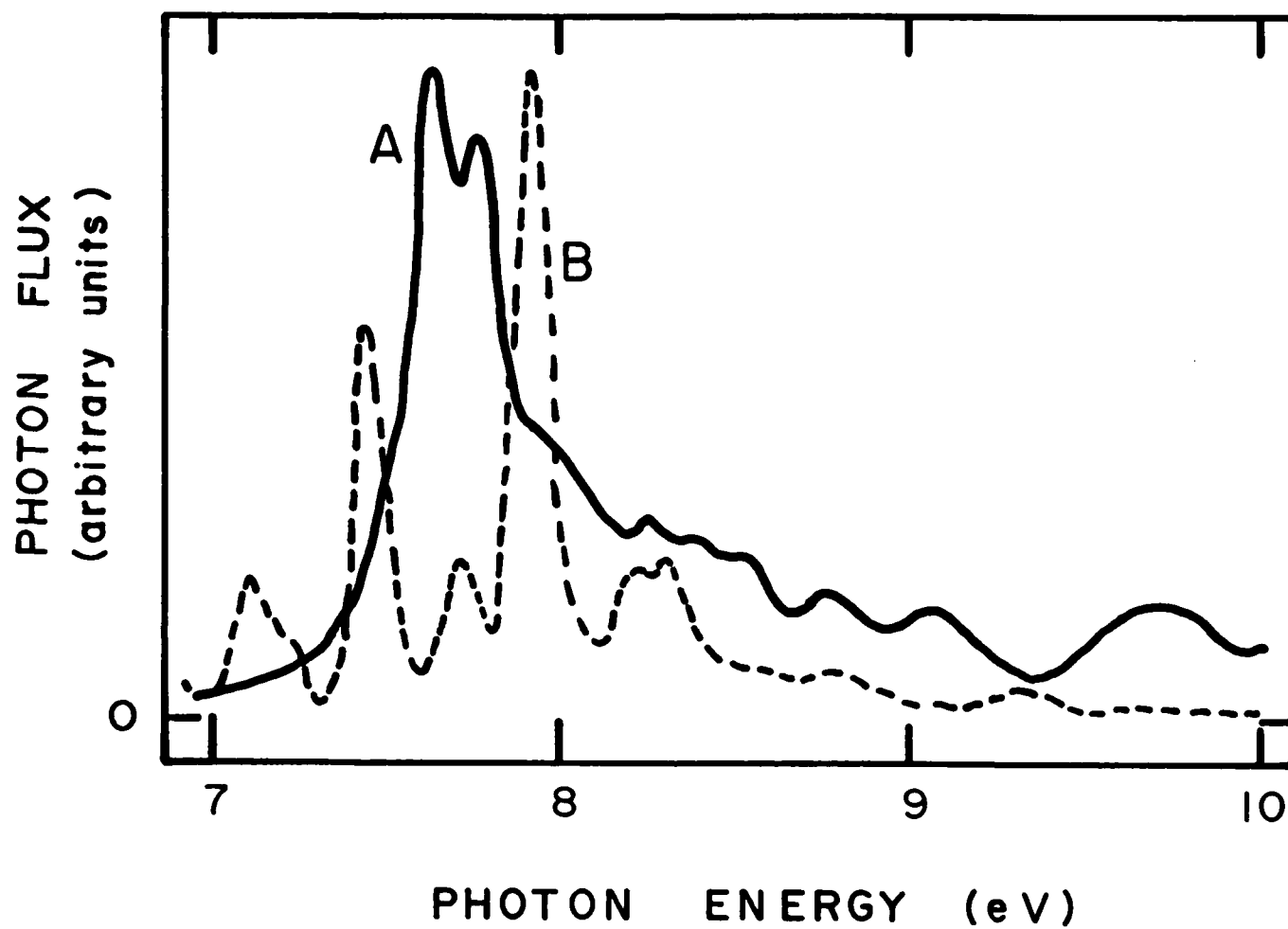


FIG. 2

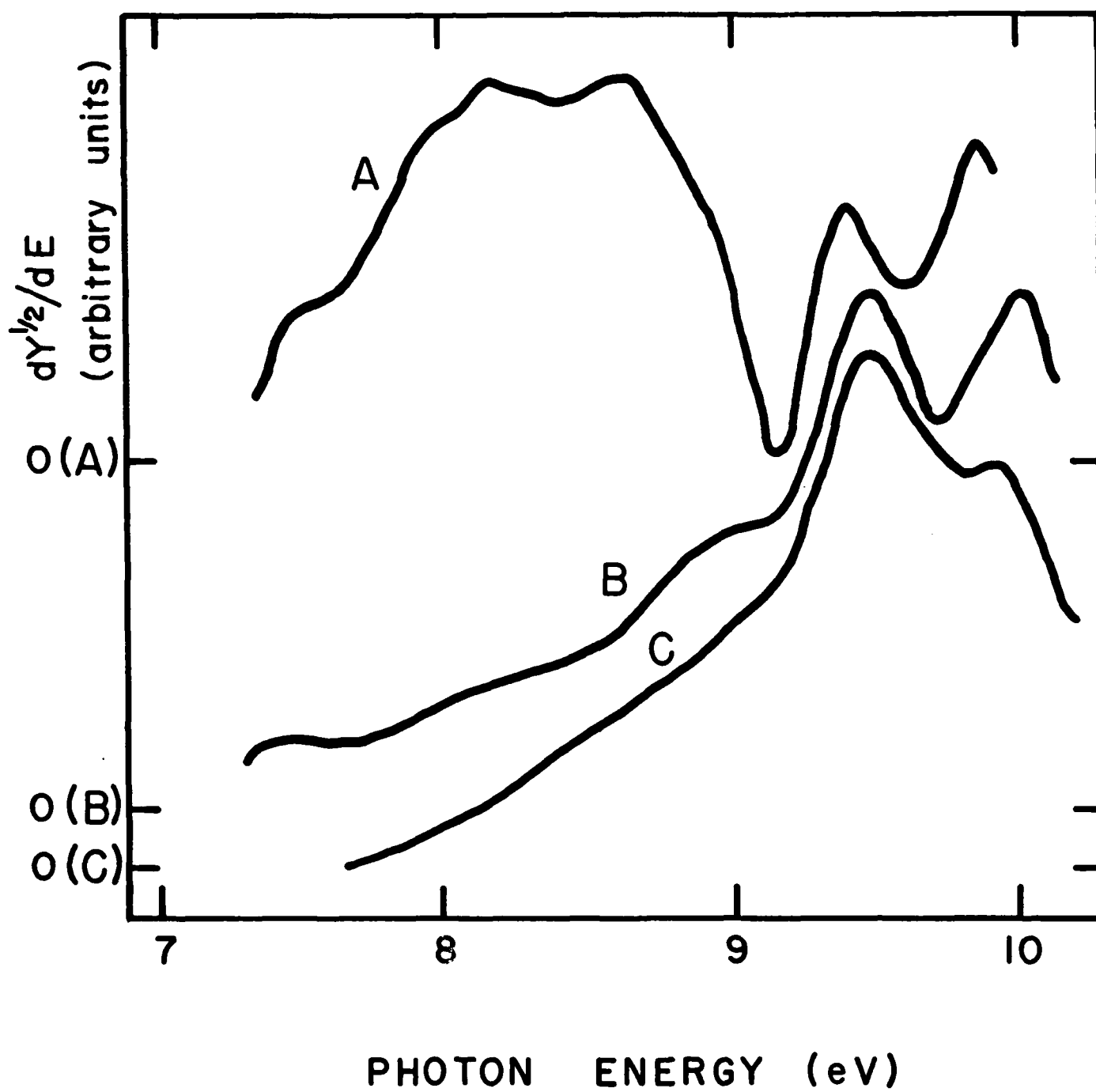


FIG. 3



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